

### REMARKS

As requested, the amendment to the specification has been resubmitted to eliminate the partially blocked and illegible material.

The claims have been amended to include the prior claim 10/17 recitation that the internal electrodes are primarily silver in the independent claims and to have claims specific to actuators and emitter (see page 1, line 8). The other changes are a matter of form only in order to provide better antecedent basis and better grammar to the claims.

The presence of internal electrode materials dispersed within a ceramic body have led to the deterioration of piezoelectric properties and poor reliability as attempts have been made to reduce the size of monolithic piezoelectric parts by reducing the thickness of the layer of the ceramic sheets which make up such parts and increasing the number of sheets. One such property is the piezoelectric d constant which relates the amount of mechanical strain produced by an applied electric field. In certain monolithic piezoelectric parts such as monolithic piezoelectric actuators and audio emitters, a high constant is required.

It is recognized in the art that when the piezoelectric ceramic is a lead perovskite type material, the piezoelectric d constant is conspicuously deteriorated when sintering in a low oxygen atmosphere because the low oxygen concentration promotes the generation of oxygen pores. When the oxygen concentration is reduced to less than 1 percent by volume or when a soft piezoelectric ceramic material with a high piezoelectric d constant is used, the deterioration is so severe that it is difficult to even form the desired monolithic piezoelectric part.

The present invention is based, in part, on the discovery that a layered article which has a plurality of piezoelectric ceramic layers and spaced internal electrode

layers disposed in the part can be fabricated without deteriorating the piezoelectric constant by co-sintering the ceramic layers and internal electrode layers in an atmosphere where the oxygen concentration is up to about 5 volume percent, but not zero percent, when the molar quantity of the A site of the perovskite has been reduced by about 0.5 to 5 mole percent from the stoichiometric amount, and the average valence of the B site component is increased to greater than 4 but less than 4.1. The fact that the deterioration of the constant can be combated is surprising and unexpected.

Claims 1, 4, 5, 7 and 14-16 were rejected under 35 U.S.C. 103 over Randall in view of Ponomarev and Horikawa '408. This rejection is respectfully traversed.

Randall teaches co-firing multi-layered piezoelectric ceramic materials with base metal electrodes. The reference acknowledges that there are significant problems with such co-firing, including, *inter alia*, firing without oxidation of the electrodes and also without reduction of the PZT or other ceramic material. See e.g. paragraph [0007]. Randall teaches that these problems can be overcome by applying a layer comprising an organic material, flux and a dopant to a piezoelectric ceramic layer, then applying a layer of a base metal powder coated with an inorganic material (and optionally a noble metal) for passivating the base metal ([0014]) in an organic material, and then continuing application of these layers until the desired structure is achieved. The resulting structure is heated to remove the organic material and then sintered at a partial oxygen pressure of about  $10^{-3}$  to  $10^{-10}$ . As the Examiner has acknowledged, Randall fails to teach or suggest using a perovskite ceramic whose A and B site molar quantities have been altered from the stoichiometric composition (i.e.,  $ABO_3$ ). Note that Randall also does not teach internal electrodes which are predominately silver but instead requires use of a base metal coated with a passivating agent.

The Ponomarev reference has been cited solely to teach that low loss hard piezoelectric ceramic materials are desired in multi-layer ceramic transformers.

Horikawa '408 relates to low loss piezoelectric ceramics for high frequency filters and isolators. The ceramic has the particular composition given at column 2, lines 50 to 55 and is formed by preparing a ceramic molded body of the ceramic which is sintered and thereafter processed for the application of electrodes. There is no teaching or suggestion in this reference of cofiring a green (unsintered) piezoelectric ceramic with internal electrodes under any conditions. In addition, there is no teaching or suggestion that the ceramic should be fired in a reduced oxygen atmosphere for any reason. Accordingly, Horikawa '408 did not have to face the problem of deterioration of the d constant.

The rejection in this case is predicated on substituting the Horikawa '408 ceramic for the Randall transformer ceramic based on the assertion that Ponomarev teaches a low loss material would be advantageous when making a transformer. But Randall did not have to be concerned with the piezoelectric d constant when making a transformer because the ceramic was stoichiometric and the electrodes were of a base metal. However, if either of these aspects are changed when making an actuator or emitter part, the effect on the constant must be taken into account. Here, the prior art discussed on page 2 of the application teaches the skilled person that piezoelectric transformer made with hard type ceramic and silver electrodes achieved good properties by increasing the A site perovskite component. It also teaches the artisan that sintering a lead perovskite piezoelectric ceramic material in a reduced oxygen atmosphere to suppress silver dispersion in the ceramic lead to significant deterioration in the piezoelectric d constant by generating oxygen pores which could even render the sintered ceramic unusable for its intended purpose. This fact is illustrated in the comparative examples of Table 3 in the present application.

The Examiner has taken issue based on the proposition that one would not expect the ceramic to have deteriorated the piezoelectric d constant because Randall clearly discloses that the piezoelectric ceramic can be sintered in a reducing atmosphere without being reduced. It is respectfully submitted that this assertion improperly equates, without any factual basis, reduction of the piezoelectric ceramic with deterioration of the piezoelectric d constant. In that connection, it is known that a piezoelectric ceramic can be sintered in a reducing atmosphere without being reduced at the same time the piezoelectric d constant deteriorates. Randall provides no support for the proposition because it is not concerned about, and has no disclosure whatsoever about, the piezoelectric d constant. Further, the proposition is contrary to the article by Moon and Jang, *Effects of Sintering Atmosphere on Densification Behavior and Piezoelectric Properties of  $Pb(Ni_{1/3}Nb_{2/3})O_3$ - $PbTiO_3$ - $PbZrO_3$  Ceramics*, J. Am. Ceram. Soc. 76[2] 549-52(1993) (See Table 1 and Fig. 4) which was submitted with the last response in this case. In this article, Moon and Jang teach that the piezoelectric constant  $d_{33}$  of the piezoelectric specimen varies in direct response to the oxygen concentration, i.e., it increases as the partial pressure of oxygen in the sintering atmosphere is increased and decreases as the partial pressure is decreased. See, e.g. particularly Table 1 and Figure 4.

The record in this case clearly establishes that those skilled in the art would expect a lead perovskite piezoelectric ceramic material to have a deteriorated piezoelectric d constant if sintered in a reduced oxygen atmosphere. The fact that the deterioration can be combated when making an actuator or emitter by employing a ceramic whose A and B site stoichiometric amounts have been altered as claimed is not taught or suggested and is surprising and unexpected.

In an Advisory Action, the Examiner commented that the deterioration effect is not being claimed. It is respectfully submitted that this approach fails to give proper

consideration to the fact that the method as claimed gives rise to an effect which is both surprising and unexpected, rendering the method unobvious.

Also, the Advisory Action asserts the reducing the lead “would compensate for oxygen pores formed during sintering in the low oxygen atmosphere of the references and thus suppress reduction in piezoelectric d constant.” But the only factual basis in the record relating to compensation in connection with silver electrodes and low oxygen is on page 2 of the instant application, and this teaches that in a transformer piezoelectric, the A site component compensates when it is increased. Decreasing the A site amount, as in the invention, is contraindicated by this art as it suggests such action would aggravate the reduction in the constant.

In light of all of the foregoing, withdrawal of this rejection is respectfully solicited.

Claims 1, 4, 5, 7-11 and 14-17 have also been rejected under 35 U.S.C. over Horikawa '328 in view of Ponomarev, Horikawa '408 and JP '433. This rejection is also respectfully traversed.

The Horikawa '328 reference has been cited to show co-sintering of a structure having laminated green piezoelectric ceramic and internal electrode layers. There is no disclosure of altering the stoichiometry of the perovskite or sintering in a reduced oxygen atmosphere. The Ponomarev and Horikawa '408 references have been discussed above.

JP '433 teaches that a hard piezoelectric ceramic having silver electrodes in which the diffusion of silver into the ceramic is reduced by sintering at an oxygen density of 1-10 percent. Note that the composition of the ceramic is stoichiometric.

This combination of references suffers from the same deficiencies discussed in the previous art rejection. The hypothesized combination would not be made since it is well known that sintering a lead perovskite piezoelectric ceramic under low oxygen conditions would result in a deterioration of the piezoelectric d constant. The Examiner has stated that JP '433 teaches that sintering in a reduced oxygen atmosphere to reduced diffusion of the silver into the ceramic still results in an excellent piezoelectric property. In response, Applicants respectfully point out that the reference teaches that this result is achieved by insuring the stoichiometric amounts are maintained, either by conducting sintering in a sealed atmosphere to prevent lead loss, or adding extra lead to make the lead content in the A site fall in a particular range. In this connection, see paragraph [0025] in either the machine translation or the more understandable translation submitted by applicants. There is no teaching or suggestion that the piezoelectric d constant deterioration can be effected by increasing the B site amount above stoichiometric, and quite to the contrary, stoichiometric values are maintained. Accordingly, the finding that increasing the average valence of the B site so that it is greater than 4 but less than 4.1 can be employed to combat the deterioration of the piezoelectric d constant is surprising and unexpected and this rejection should be withdrawn.

Claim 6 and 21 were rejected under 35 U.S.C. § 103 over the same combination of references in further combination with Feltz. This additional reference has been cited only to teach that it is possible to do partial substitution of the ions in the B position of the perovskite but not to overcome the deficiencies in the prior combinations of references discussed. Accordingly, the further combination of these references with Feltz cannot render the invention obvious.

Claims 12 and 13 were rejected under 35 U.S.C. 103 over the references applied to claim 11 and in further combination with JP '035. The additional Japanese

reference has been cited only with regard to the internal electrodes and not to overcome the basic deficiencies in the base combination of references. It does not do so and withdrawal of this rejection is respectfully solicited.

In light of all of the foregoing, it is respectfully submitted that this application is now in condition to be allowed and the early issuance of a Notice of Allowance is respectfully requested.

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